Light Transport and Localization in Diffusive Random Lasers

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Abstract.
We develop an analytical theory for diffusive random lasers by coupling the transport theory of the disordered medium to the semiclassical laser rate equations, accounting for (coherent) stimulated and (incoherent) spontaneous emission. From the causality of wave propagation in an amplifying, diffusive medium we derive a novel length scale which we identify with the average mode radius of the lasing quasi-modes. We show further that loss at the surface of the laser-active medium is crucial in order to stabilize a stationary lasing state. The solution of the transport theory of random lasers for a layer geometry with appropriate surface boundary conditions yields the spatial profile of the light intensity and of the population inversion. The dependence of the intensity correlation length on the pump rate is in qualitative agreement with experimental and numerical findings.

1. Introduction

A random laser is a system formed by randomly distributed scatterers embedded in a host medium where either scatterer or host medium or both provide optical gain through stimulated emission [1]. Recent observations of random lasing in a wide variety of systems, like powders of semiconductor nanoparticles [2, 3, 4, 5], organic dyes in strongly scattering media [6, 7, 8], organic films or nanofibers [9, 10, 11] and ceramics [12], have triggered a rapidly growing interest. For reviews with comprehensive lists of references see [13, 14]. Random lasers share some properties with conventional lasers, like threshold behavior [4], narrow spectral lines [15], or photon statistics [16, 17], but also exhibit distinctly different properties like multidirectional emission. Coherent feedback has unambiguously been demonstrated to be present in strongly disordered random lasers [16]. It requires the light to be sufficiently confined in the random system. While spatially confined regions from which the laser emission takes place have been observed experimentally [15, 13], the physical origin of coherent feedback, of localized quasimodes and the dependence of their size on the pump rate have remained controversial. The possible theoretical explanations range from preformed random microresonators [18] to multiple random scattering (diffusion) [19], possibly enhanced by self-interference [15] of waves and the resulting onset of Anderson localization (AL) [20]. Conversely, it is an interesting fundamental question how AL of light, which has been understood[21, 22] as a consequence of self-interference, is influenced by the coherent, stimulated amplification in the lasing state. Problem of the intensity distribution in a diffusive random laser has been attacked theoretically

by phenomenolocical diffusion models [23] and numerical calculations in one [24, 25] and two spatial dimensions [26, 27, 28, 19]. However, the experimentally observed decrease of the lasing spot size with increasing pump rate, has not been explained so far.

In this paper, we address the question of the size of lasing spots in diffusive random lasers by an analytical transport theory. We begin the analysis in section 2 by an outline of the transport theory of light in a disordered medium with linear gain (i.e., constant-in-time amplification rate), including self-interference (so-called Cooperon) contributions. By a phenomenological analysis we show that causality implies in the presence of linear gain a novel length scale which is to be identified by the average radius (spot size) of a lasing mode, R_s. In section 3 we extend the theory for a system with linear gain to a transport theory including non-linear gain due to stationary lasing above threshold. Observing that a stationary lasing state is possible only if the amplification in the medium is compensated by loss at the surface of the system, we consider a model for a finite-size random laser with infinite extension in the (x,y) plane, but finite, constant thickness in the z direction, a geometry relevant for many experimental systems [3, 4, 15, 16]. Coupling the diffusive transport theory to the rate equations of a four-level laser in the stationary state we derive an analytical expression for the intensity correlation length ξ to be identified with the average lasing spot size, R_s. Due to the surface boundary conditions the spatial extention of a lasing spot in the (x,y) plane obtains a z-dependent profile , $\xi(L)$. We also analyse its dependence on the pump rate. The conclusions are drawn in section 4. on the pump rate and on the depth of the mode along the z direction.

2. Transport Theory for a Diffusive, Linear-gain Medium and Causality

The propagation of light is described by its wave equation. Neglecting the polarization degree of freedom we consider in the following the scalar wave equation for the field Ψ. It reads,

$$\frac{\omega^2}{c^2}\,\epsilon(\vec{r})\Psi_\omega(\vec{r}) + \nabla^2\Psi_\omega(\vec{r}) = -i\omega\frac{4\pi}{c^2}j_\omega(\vec{r})\ , \tag{1}$$

where c denotes the vacuum speed of light and $j_\omega(\vec{r})$ an external current source. The dielectric constant is $\epsilon(\vec{r}) = \epsilon_b + \Delta\epsilon\,V(\vec{r})$, where the dielectric contrast between the background, ϵ_b, and the scatterers, ϵ_s, has been defined as $\Delta\epsilon = \epsilon_s - \epsilon_b$. The spatial arrangement of the scatterers is described through the function $V(\vec{r}) = \sum_{\vec{R}} S_{\vec{R}}\,(\vec{r} - \vec{R})$, with $S_{\vec{R}}\,(\vec{r})$ a localized shape function at random locations $\vec{R}$. Linear gain (absorption) is described by a temporally constant, negative (positive) imaginary part of ϵ_b and/or ϵ_s.

In Refs. [29, 30, 31] we have developed a theory for light transport in disordered media with linear gain or absorption. It results in an energy-density correlation function $P_{\mathrm{E}}^\omega(\vec{r} - \vec{r}', t - t')$, which describes how the energy density of the light field with frequency ω propagates diffusively between two points in space and time, $(\vec{r}, t)$, $(\vec{r}', t')$. The Fourier transform of the energy-density correlation function $P_{\mathrm{E}}^\omega(Q, \Omega)$ is obtained as

$$P_{\mathrm{E}}^\omega(Q, \Omega) = \frac{N_P}{\Omega + iQ^2D + i\xi_a^{-2}D} \tag{2}$$

where the expression for the coefficient N_P is given explicitly in Ref. [30], but is not relevant for the present purpose. The denominator of Eq. (2) exhibits the expected

diffusion pole structure with the diffusion coefficient D. In addition, in the case of a non-conserving medium, i.e. net absorption (gain), there appears the (purely imaginary) term $i\gamma_a = i\xi_a^{-2}D$, which has a positive (negative) imaginary part and does not vanish in the hydrodynamic limit, $\Omega \to 0$, $Q \to 0$. The self-consistent solution of the transport theory including self-interference of waves (Cooperon contributions) (see Refs. [30, 31]) shows that in the presence of absorption or gain the diffusion coefficient D cannot vanish and is in general complex. Hence, truly Anderson localized modes do not exist in this case.

For the case of absorption ($\gamma_a > 0$) it is seen by Fourier transforming Eq. (2) w.r.t. time, $P_{\mathrm{E}}^{\omega}(Q,t) = iN_P\, e^{-(Q^2 D + \gamma_a)t}$, that $Re\gamma_a$ represents the loss rate of the photonic energy density due to absorption in the medium. Fourier transforming, on the other hand, Eq. (2) w.r.t. space in the stationary limit ($\Omega \to 0$)) shows that $\xi_a = Re\sqrt{(\gamma/D)}$ is the length scale over which the energy density of diffusive modes is correlated in the lossy medium.

For the case of linear gain ($\gamma_a < 0$) the wave equation predicts an unlimited growth of the field amplitude and, hence, of the energy density. This means that a stationary lasing state is not possible in this case and, therefore, the limit $\Omega \to 0$ must strictly not be taken in Eq. (2). Such a behavior of linear gain is expected only during the exponential intensity growth shortly after the onset of lasing. A complete theory of random lasing must, therefore, take into account either the full temporal dynamics of the system, or in a stationary state additional surface loss effects must compesate for the gain in the medium (see section 3). Nevertheless, we can extract a characteristic size of a stationary lasing spot from this theory by requiring that the stationary lasing state has been reached *locally*, i.e. within a finite subvolume of the system: Causality requires that the pole of $P_E^{\omega}(Q,\Omega)$, Eq. (2), as a function of Ω resides in the lower complex Ω half-plane. For $\gamma_a < 0$ this is possible only if all the diffusive modes allowed inside a given lasing spot have a wavenumber $Q > Q_{min} = \sqrt{Re(-\gamma_a/D)}$. This, in turn, requires that the spot size is

$$R_s = \frac{2\pi}{Q_{min}} = \frac{2\pi}{\sqrt{Re(-\gamma_a/D)}} \ . \tag{3}$$

It is the characteristic, maximal size of a spatial region over which diffusive modes can be causally correlated in the stationary lasing state. We conjecture that, hence, this size is to be identified with the lasing spot size observed experimentally [15, 13] in diffusive random lasers. Since according to the microscopic theory [30] the growth rate $(-\gamma_a)$ is, for small linear gain, proportional to the avergage gain in the medium, $\gamma_a \propto Im\epsilon(\vec{r})$, we predict the spot size to be inversely proportional to the gain.

More generally, despite the fact that the linear gain assumption is not suited to describe stationary lasing, it can be used to estimate the laser threshold, i.e. the critical pump rate for lasing. Amazingly, this is a rather general remark. For example, in a simpler system of a single microsphere with gain, it has been shown [32], that the scattering coefficients calculated within linear response lose their causality just at the point where the sphere crosses its lasing threshold. Applied to our random laser system, this means that the threshold for lasing within a spot of size R_s is reached when the transport coefficient $-\gamma$, determined by the pump rate via the microscopic transport theory Refs. [30, 31], reaches the value given by Eq. (3)

In Fig. 1 we show the numerical evaluation of the spot size R_s as a function of increasing $Im\epsilon_s$ for typical parameters, as given in the figure caption. The imaginary part of the dielectric constant is a measure of external pumping, since the gain is
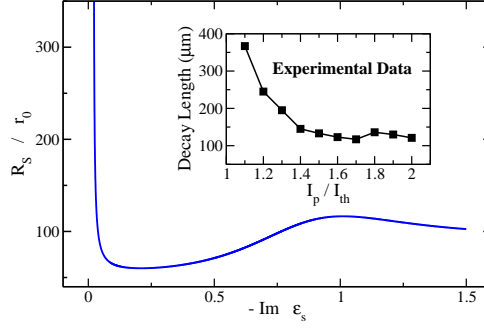
Figure 1. The spot size R_s, Eq. (3) in units of the scatterer radius r_0, as obtained by causality considerations (see text) as a function of the imaginary part of the dielectric constant of the scatterers. The parameter values used are $\epsilon_b = 1$, $Re\epsilon_s = 10$, scatterer filling fraction $\nu = 30\%$, light frequency $\omega/\omega_0 = 2.5$. The ligth frequency is $\omega_0 = 2\pi c/r_0$ where c is the vacuum speed of light. The data in the inset are taken from Ref. [13] and refer to the spot size of the modes.

given by the population inversion of the laser. Therefore, larger pumping yields higher inversion and leads to a larger $Im\epsilon_s$. The calculated spot size displays a qualitative agreement with the experimental data [13] (spot size vs. pump intensity / threshold intensity) shown in the inset.

3. Tranport Theory of Random Lasing

As remarked in section II, a stationary lasing state in a homogenaously pumped system is possible only if the system is finite, so that surface loss effects can compensate the gain in the medium. To avoid the causality problem, we consider here a three-dimensional random laser model with a homogeneously pumped, active medium which extends infinitely in the (x,y) plane, but has a finite, constant thickness d in the z direction. The laser-active material is described by the semi-classical laser rate equations, and the light intensity transport by a diffusion equation. In particular, the rate equations for a four-level laser are

$$\frac{\partial N_3}{\partial t} = \frac{N_0}{\tau_P} - \frac{N_3}{\tau_{32}} \tag{4}$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} - \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 - \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} \tag{5}$$

$$\frac{\partial N_1}{\partial t} = \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 + \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} - \frac{N_1}{\tau_{10}} \tag{6}$$

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}} - \frac{N_0}{\tau_P} \tag{7}$$

$$N_{tot} = N_0 + N_1 + N_2 + N_3, \tag{8}$$

where $N_i = N_i(\vec{r}, t)$, $i = 0, 1, 2, 3$ are the population number densities of the corresponding electron level ($i \in \{1\ldots4\}$), N_{tot} is the total number of electrons participating in the lasing process, $\gamma_{ij} \equiv 1/\tau_{ij}$ are the transition rates from level i to j, and γ_{nr} is the non-radiative decay rate of the laser level 2. $\gamma_P \equiv 1/\tau_P$ is the transition rate due to homogeneous, constant, external pumping. Furthermore

$n_{ph} \equiv N_{ph}/N_{tot}$ is the photon number density, normalized to N_{tot}. In the stationary limit (i.e. $\partial_t N_i = 0$), the above system of equations can be solved for the population inversion $n_2 = N_2/N_{tot}$ to yield (γ_{32} and γ_{10} assumed to be large compared to all other rates)

$$n_2 = \frac{\gamma_P}{\gamma_P + \gamma_{nr} + \gamma_{21}(n_{ph} + 1)}, \tag{9}$$

The photon number density (light intensity), normalized to N_{tot}, $n_{ph} = N_{ph}/N_{tot}$, obeys the diffusion equation [23],

$$\partial_t n_{ph} = D_0 \nabla^2 n_{ph} + \gamma_{21}(n_{ph} + 1)n_2, \tag{10}$$

where the last term on the r.h.s. describes the intensity increase due to stimulated and spontaneous emission, as described by the semi-classical laser rate equations. Since in the slab geometry ensemble-averaged quantities are translationally invariant in the (x, y) plane, but not along the z direction, a Fourier representation in the (x, y) plane in terms of $n_{ph}(\vec{Q}_{||}, z)$, $n_2(\vec{Q}_{||}, z)$ is convenient,

$$\partial_t n_{ph} = - D_0 Q_{||}^2 n_{ph} + D_0 \partial_z^2 n_{ph} \tag{11}$$
$$+ \gamma_{21} \int \frac{d^2 Q_{||}'}{(2\pi)^2} n_{ph}(\vec{Q}_{||} - \vec{Q}_{||}', z) n_2(\vec{Q}_{||}', z) + \gamma_{21} n_2$$

We now seek the photon density response function $P(\vec{Q}_{||}, z, \Omega)$, which describes the response of the photon density, $n_p h$, to the distribution of the population inversion, n_2, in order to determine the transport coefficients. In the stationary case ($\partial_t n_{ph} = 0$) and in the long-wavelength limit along the (x, y) plane ($Q_{||} \to 0$), the z derivative in Eq. (11) can be expressed without derivatives in terms of n_{ph} and n_2 only. Plugging this back into Eq. (11) yields,

$$\left[\partial_t + D_0 Q_{||}^2 + \frac{\gamma_{21} n_2}{n_{ph}} \right] n_{ph}(\vec{Q}_{|||}, z, t) = \gamma_{21} n_2(\vec{Q}_{|||}, z, t) \tag{12}$$

and, hence, after Fourier transform w.r.t. time, the diffusion form of the density response function,

$$P_E(\vec{Q}_{||}, z, \Omega) = \frac{i\gamma_{21}}{\Omega + iQ_{||}^2 D_0 + i\xi^{-2} D_0}, \tag{13}$$

where from Eq. (12) the correlation length ξ is defined as the real, positive quantity,

$$\xi = \sqrt{\frac{D_0}{\gamma_{21}} \frac{n_{ph}}{n_2}}. \tag{14}$$

As seen from Eq. (13) the pole structure of P_E in this finite-size, diffusive system is perfectly causal. The square of the correlation length ξ remains positive, indicating an effective loss out of a given $Q_{||}$ mode. This is due to the loss of intensity at the surfaces. Additionally, the mass term becomes less and less significant as the laser intensity in the sample builds up, because the relative population inversion clearly obeys $n_2 \leq 1$ whereas the relative photon number is not restricted.

Since for homogeneous pumping the averaged photon density does not depend on x or y, Eq. (11) simplifies in the stationary limit to

$$D_0 \partial_z^2 n_{ph} = -\gamma_{21}(n_{ph} + 1)n_2 \tag{15}$$
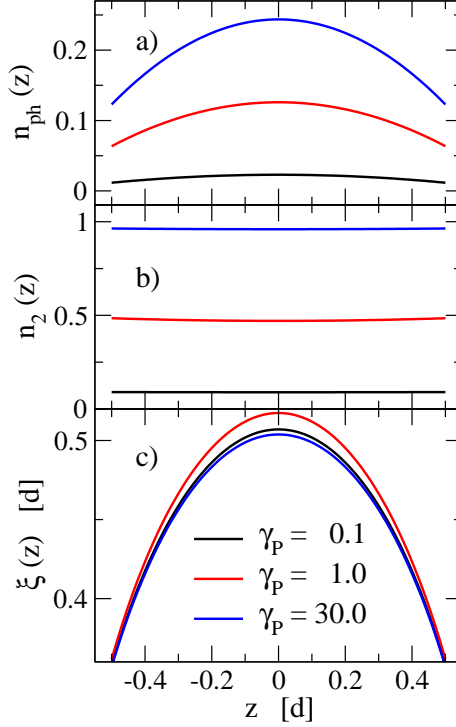
Figure 2. The following quantities are shown as a function of z for different values of the pump rate γ_P: a) the photon number, which increases monotonically with increasing pump rate and has its maximum in the film center ($z = 0$); b) the population inversion, which is inverse proportional to n_{ph}, see Eq. (9); c) the correlation length (spot size), which clearly behaves non-monotonically with increasing pumping. For all panels the diffusion constant is $D_0 = 1d^2\gamma_{21}$.

and $n_{ph}(z)$ is finally determined via Eq. (9) by the regular differential equation,

$$\partial_z^2 n_{ph}(z) = -\frac{\gamma_{21}}{D_0} \; \frac{(\gamma_P/\gamma_{21})}{1 + \frac{(\gamma_P/\gamma_{21})}{n_{ph}(z)+1}} \; . \tag{16}$$

Eqs. (16) (9) and (14) comprise the complete description of the spatial photon density profile perpendicular to the lasing film and the intensity correlation length (spot size) parallel to the film.

Numerical evaluations of Eqs. (16), (14) and (9) are shown in Figs. 2 and 3. In Fig. 2 the photon number $n_{ph}(z)$, population inversion $n_2(z)$ and correlation length $\xi(z)$ are shown as a function of z for different values of external pumping, characterized by the pumping rate γ_P. The value of the diffusion constant was chosen to be $D_0 = 1d^2\gamma_{21}$, where d is the width of the film. In panel a) of Fig. 2 the photon number displays a monotonically increasing behavior with increasing pumping. The maximum of the intensity resides in the center of film ($z = 0$), since this is the position farthest from the boundaries, and therefore with lowest loss of intensity. The population inversion, Eq. (9), behaves inverse to $n_{ph}(z)$, see Eq. (9). In contrast to this rather expected behavior, the correlation length $\xi(z)$ as given by Eq. (14) exhibits a non-monotonic behavior with increasing pumping. For pumping rates $\gamma_P < \gamma_{21}$ the
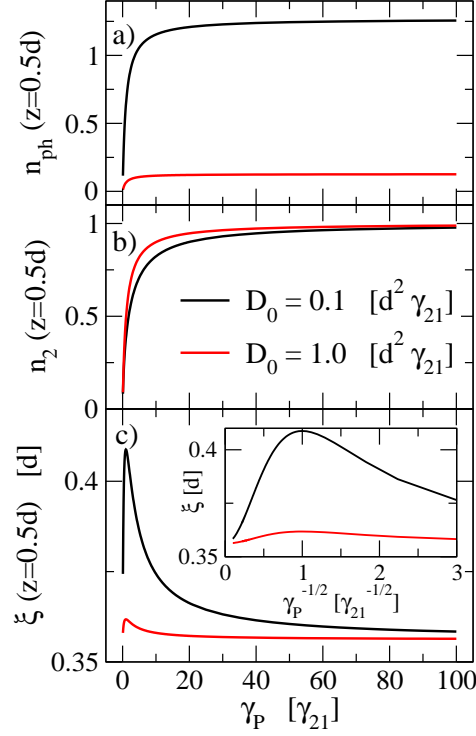
Figure 3. The figure shows the following quantities as a function of the pumping rate P at the film surface ($z = \pm 0.5d$): a) the photon number which displaying saturation for strong pumping; b) the population inversion, also saturating; c) the correlation length (spot size) showing non-monotonic behavior. The inset of panel c) displays the $1/\sqrt{\gamma_P}$ dependence of the intensity correlation length ξ on the pump rate above threshold ($\gamma_P > 1$) (see discussion in the text).

correlation length increases but for pumping rates $\gamma_P > \gamma_{21}$, ξ is decreasing. The equality between γ_P and γ_{21} marks the situation where electrons are as fast excited into the upper laser level as they relaxate to lower levels. Therefore this characterizes the lasing threshold. Available experimental data [15, 13] also report a decreasing behavior of the spot size above threshold. Measurements of the intensity correlation length below threshold have not yet been reported.

The same quantities are shown Fig. 3 as a function of external pumping γ_P at the surface of the random laser. Photon number and population inversion both display saturation behavior. Panel c) of Fig. 3, however, exhibits the non-monotonic behavior of the correlation length. This plot is to be directly compared to experimental data [13], as e.g. shown in the inset of Fig. 2. There is a good qualitative and even quantitative agreement between calculated and measured spot size. The inset of panel c) exhibits shows that the dependence of the spot size ξ on the pump rate γ_P above threshold (γ_P) is predicted to be

$$\xi(\gamma_P) = \xi(\infty) + \alpha/\sqrt{\gamma_P} \tag{17}$$

with a proportionality constant α. This result is open for further experimental tests.

4. Conclusion

We have discussed how a linear response theory for light transport, including self-interference effects, in disordered media with linear gain predicts threshold behavior of the intensity. Even more interestingly it also predicts a characteritic, average radius of lasing modes, dictated by causality. We identify this length scale with the spot size of the random laser as measured in experiments [15, 13] and find qualitatively good agreement. Further, we have proposed an analytical transport theory for random lasing in finite systems. A finite system size is necessary for surface loss to compensate the gain in the medium and, hence, to stabilize a stationary lasing state. In particular, we consider a slab geometry where in the medium the light intensity propagates diffusively and the loss through the surfaces is included by appropriate boundary conditions. The theory allows for the first time for an analytical calculation of the intensity correlation length of this system, describing the spatial extent of a mode (spot size). The spot size is predicted to behave non-monotonously as a function of external pumping, i.e. increasing below and decreasing above the laser threshold. A comparison with experiments reveals qualitativly good agreement. Our prediction of its functional dependence on the pump rate is open to further experimental tests.

Our future work will include solving the semi-analytical light transport theory with self-interference contributions when the system is self-consistently coupled to the laser rate equations.

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